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National and Regional Trends in VOC (Volatile Organic Compounds) and NOx Emissions from 1900 to 1987

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16. ABSTRACT The paper gives estimates of historic emissions of VOC and NCx for each state of the contiguous U.S. The emissions were estimated for anthropogenic source categories which included transportation sources, external combustion sources, industrial processes, solid waste disposal, and miscellaneous other sources. From 1900 to the present, NCx emissions have increased steadily throughout the U.S. VOC emissions have historically been higher but, in recent years, the total emissions are nearly the same as for NCx. The reasons for these changes and source category contributions are presented. As a result of these changes, the ratio of VOC/NCx has decreased from 5.1 to 1.2 on a national basis from 1900 to 1985, and from about 20 to less than 2 in certain states. The ratio of VOC/NCx emissions is important in modeling photochemical atmospheric reactions, especially ozone. This paper presents a basis for understanding the temporal and spatial emissions trends.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group	
Pollution	Combustion	Pollution Control	13B	21B
Organic Compounds	Industrial Processes	Stationary Sources	07C	
Volatility		Volatile Organic Compounds (VOCs)	20M	13H
Nitrogen Oxides	Waste Disposal		07B	
Sulfur Dioxide				
Emission			14G	
Transportation			15E	

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National and Regional Trends in VOC and NO_x Emissions From 1900 to 1987

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INTRODUCTION

Volatile organic compounds (VOCs) and oxides of nitrogen (NO_x) are considered important to the formation of photochemical smog and ozone. While sulfur dioxide (SO_2) and NO_x have long been considered primary precursors of acid precipitation, research is showing that VOCs are also an important constituent in many atmospheric chemical reactions. The historic trend of VOC emissions is important to understanding the long term trend in ozone and smog formation.

METHOD

Annual estimates of emissions of VOCs are presented for each of the contiguous 48 states and the District of Columbia. Emissions were estimated using a detailed methodology for every fifth year from 1900 to 1985. The state level and the 5-year interval were selected to produce estimates comparable to the historic estimates of SO_2 and NO_x provided by Gschwandtner et al.[1] The VOC emissions are consistent in methodology from year to year and state to state. In addition, the total VOC emissions of all states are identical to the national emissions reported by the U.S. EPA for common years in the period from 1940 to 1985.[2] Annual emissions were estimated by apportioning the national estimates for every fifth year according to yearly activity indicators representing the major source categories.[3]

The major source categories are:

1. Transportation sources
2. External fuel combustion sources
3. Industrial processes
4. Solid waste disposal
5. Miscellaneous other sources

Values were calculated by aggregating the emissions estimated for smaller categories comprising each major category. These subcategories represent sources for which emission factors and activity indicators could be obtained directly from the U.S. EPA national trends estimates.[2] Collectively, these source categories are considered to account for all anthropogenic emissions.

RESULTS

Figure 1 shows the total VOC emissions for the contiguous United States estimated for each study year and interpolated for the intervening years. The total VOC emissions have increased over the study period with peaks around 1930 and 1970. In 1930, the emissions were higher due mostly to a larger-than-usual number of wildfires, especially in the southeastern and south central states. By 1970, emissions had increased as a result of generally greater activity, especially in the transportation and industrial process categories. After 1970, the total VOC emissions appear to decrease primarily as a result of improved emission control efficiencies on automobiles and many industrial processes, as reflected in the emission factors derived for many of these source categories.

In 1900, transportation sources contributed 2 percent of the total emissions. By 1960, these sources contributed nearly half of the total VOC emissions. Also, the percentage contribution of industrial processes increased from about 3 percent in 1900 to 26 percent in 1960 and 39 percent in 1980. External fuel combustion was a major category from 1900 to 1940, but the percentage contribution has decreased since then. Similarly, the contribution by miscellaneous other sources has also decreased with time, primarily due to the success of fire prevention programs. The percentage contribution of solid waste disposal appears to have remained nearly constant over time.

Figure 2 shows the overall trend in VOC emissions from each major category for the contiguous United States. The figure shows the declining contribution of external fuel combustion sources and the increasing contribution of transportation sources and industrial processes. In the 1920s and 1930s, wildfires contributed significantly to the total VOC emissions which results in the intermediate peak. The contribution of wildfires may also have been significant in earlier years, although exact statistics are not available. From 1970 to 1985, the emissions of many source categories decreased, despite increased activity, as a result of implementing emission controls as reflected in the estimated emission factors.

COMPARISON TO SO_2 AND NO_x EMISSIONS

Figure 3 shows the total VOC emissions estimated in this study and the total yearly SO_2 and NO_x emissions estimated by Gschwandtner et al. for 1900 to 1980.[1] The figure shows that the emissions of each of these pollutants have changed over time, but to different degrees. NO_x emissions have increased nearly steadily since 1900, while SO_2 emissions have gradually increased from 1900 to 1980 with considerable changes during certain shorter periods of time. VOC emissions have also increased, but the rate of increase appears to be less than that of NO_x and more than SO_2, at least on the national level.

RATIO OF VOC TO NO_x EMISSIONS

Since the 1940s, transportation and industrial process have contributed most of the VOC emissions. Transportation sources are also major contributors of NO_x. The ratio of VOC to NO_x emissions on the national scale has decreased from 5.1 (based as the median VOC emission estimate in 1900) to 1.2 in 1985, despite an increase in the total emissions of each pollutant. The ratio of VOC to NO_x is an important tool in modeling photochemical pollution such as ozone and smog, and in determining emission reductions necessary to attain national ambient standards for ozone.

Table I presents the estimated total emissions of VOC on the state level for every tenth year from 1900 to 1980 and 1985. Estimates for every fifth year are available on the state level from Gschwandtner and Wagner.[3]

Table II presents the total emissions of NO_x on the state level for the same years. Estimates for every fifth year are available on the state level from Gschwandtner et al.[1] from 1900 to 1980. For 1985, the emission estimates were obtained by Zimmerman et al.[4]

Table III presents the ratio of VOC emissions to NO_x emissions for each state. For states such as Oregon, Washington, and Idaho where wood burning was the predominant source, the VOC to NO_x ratio has decreased from about 20 to about 2. For industrial states such as New York, Ohio, and Illinois, the ratio has changed the least due to a mix of predominant sources that collectively emit VOC and NO_x in comparable amounts.

The ratio of VOC to SO_2, however, has remained about equal to unity over the study period, even though these pollutants are emitted by different sources and to different degrees. Figure 4 shows graphically the historic ratios of VOC emissions to SO_2 and NO_x emissions.

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Table 1. VOC emissions from 1900 to 1985
(1000 metric tons)

STATE	1900	1910	1920	1930	1940	1950	1960	1970	1980	1985
ALABAMA	399	384	367	517	400	410	452	490	441	431
ARIZONA	54	58	63	81	93	141	215	274	288	276
ARKANSAS	351	336	272	872	375	281	307	308	284	279
CALIFORNIA	555	564	680	923	1073	1656	2211	2771	2418	2279
COLORADO	83	91	105	131	142	202	266	314	304	286
CONNECTICUT	103	105	116	158	179	212	266	349	249	232
DELAWARE	31	28	28	36	36	43	59	66	56	53
D.C.	8	9	17	35	59	74	70	67	39	36
FLORIDA	387	379	314	2239	911	743	780	882	989	933
GEORGIA	614	584	468	1003	775	689	551	670	582	561
IDAHO	130	130	96	99	107	123	144	132	108	108
ILLINOIS	350	406	442	656	773	964	1127	1282	919	851
INDIANA	241	281	302	385	404	545	647	746	643	593
IOWA	105	114	140	221	245	328	364	420	259	243
KANSAS	86	122	131	216	218	302	335	365	290	271
KENTUCKY	210	205	206	317	302	344	369	432	427	415
LOUISIANA	448	444	343	471	341	417	534	630	590	555
MAINE	179	170	167	163	153	150	152	150	136	138
MARYLAND	165	160	164	196	205	268	352	470	387	369
MASSACHUSETTS	280	281	314	395	409	462	541	598	457	436
MICHIGAN	354	355	392	568	610	770	926	1178	815	769
MINNESOTA	307	310	287	346	344	376	460	489	388	369
MISSISSIPPI	449	429	362	493	815	425	325	329	277	266
MISSOURI	394	398	354	405	451	565	631	630	500	481
MONTANA	107	114	105	114	132	158	167	156	134	129
NEBRASKA	64	73	90	126	123	184	210	232	151	141
NEVADA	31	36	39	50	58	76	84	96	101	98
NEW HAMPSHIRE	75	73	72	75	73	73	78	95	90	88
NEW JERSEY	211	237	276	442	458	560	706	847	640	593
NEW MEXICO	81	85	85	94	108	154	194	200	171	161
NEW YORK	462	527	648	974	1034	1316	1530	1755	1198	1133
NORTH CAROLINA	571	533	491	520	518	574	577	655	573	557
NORTH DAKOTA	27	39	52	61	66	106	124	127	105	98
OHIO	374	402	484	657	733	956	1141	1279	941	876
OKLAHOMA	124	151	189	540	632	454	414	451	428	409
OREGON	465	433	381	371	344	336	340	350	340	330
PENNSYLVANIA	633	684	749	915	981	1184	1316	1429	1071	1013
RHODE ISLAND	31	34	39	58	62	70	85	101	69	65
SOUTH CAROLINA	354	332	295	893	387	304	310	390	323	312
SOUTH DAKOTA	38	44	54	70	77	103	112	118	86	82
TENNESSEE	424	399	365	459	460	462	465	509	469	454
TEXAS	393	427	463	948	979	1344	1669	1881	2203	2096
UTAH	42	48	55	66	78	126	152	165	165	153
VERMONT	60	58	57	57	56	54	53	58	54	53
VIRGINIA	429	407	389	485	384	451	515	603	505	483
WASHINGTON	363	352	346	359	331	400	428	518	438	426
WEST VIRGINIA	124	133	140	200	175	215	210	214	183	177
WISCONSIN	312	306	362	430	366	425	479	536	420	403
WYOMING	36	41	46	58	72	99	119	115	120	111
TOTAL	12113	12311	12405	19949	18105	20674	23560	26920	22828	21674

Table II. NOx emissions from 1900 to 1985
(1000 metric tons)

STATE	1900	1910	1920	1930	1940	1950	1960	1970	1980	1985
ALABAMA	83	98	111	101	94	140	238	384	462	386
ARIZONA	3	5	7	29	34	55	50	141	296	202
ARKANSAS	143	149	161	137	72	116	105	159	201	196
CALIFORNIA	44	73	112	678	568	712	646	1266	1274	1024
COLORADO	24	40	47	49	45	74	67	151	281	240
CONNECTICUT	30	45	54	56	62	82	74	168	129	105
DELAWARE	3	3	4	6	8	17	16	48	54	56
D.C.	6	9	10	15	19	28	26	42	20	17
FLORIDA	48	54	63	326	164	196	178	460	637	561
GEORGIA	65	76	85	134	119	180	163	354	508	486
IDAHO	6	10	13	15	16	29	26	51	101	72
ILLINOIS	187	302	365	368	401	565	513	953	923	800
INDIANA	66	121	174	201	207	278	252	601	777	725
IOWA	52	75	86	82	90	152	138	255	283	218
KANSAS	30	76	63	123	110	188	170	297	407	365
KENTUCKY	34	49	62	84	89	134	122	413	513	428
LOUISIANA	39	49	80	246	188	303	275	780	798	628
MAINE	22	34	40	32	33	35	32	59	56	55
MARYLAND	33	46	53	83	93	105	95	254	258	232
MASSACHUSETTS	70	101	119	146	137	152	138	270	242	228
MICHIGAN	67	112	164	209	222	319	290	725	660	542
MINNESOTA	44	67	85	83	84	122	111	254	280	293
MISSISSIPPI	44	53	60	66	130	124	113	226	250	197
MISSOURI	73	103	120	135	145	208	188	384	491	441
MONTANA	19	27	33	39	45	67	61	74	118	128
NEBRASKA	26	37	43	42	40	70	63	133	156	136
NEVADA	2	5	6	36	26	34	31	43	100	93
NEW HAMPSHIRE	17	25	29	19	21	19	17	47	50	45
NEW JERSEY	84	142	181	344	319	291	264	435	373	304
NEW MEXICO	5	9	13	16	29	83	76	231	297	234
NEW YORK	228	352	427	511	500	607	551	847	643	515
NORTH CAROLINA	30	40	56	80	93	176	160	457	508	412
NORTH DAKOTA	6	15	24	20	19	36	33	75	128	151
OHIO	157	283	351	378	395	560	508	1018	1071	838
OKLAHOMA	12	39	99	296	229	216	196	335	441	326
OREGON	18	23	27	85	73	104	95	141	183	140
PENNSYLVANIA	250	502	621	570	529	670	607	965	1063	788
RHODE ISLAND	19	28	31	31	32	29	26	42	33	25
SOUTH CAROLINA	20	26	34	120	65	82	74	200	246	213
SOUTH DAKOTA	8	13	16	18	20	29	26	53	71	60
TENNESSEE	38	50	61	88	115	160	145	407	527	421
TEXAS	54	70	108	675	703	1076	976	1941	2312	2039
UTAH	6	12	16	22	26	67	61	82	159	140
VERMONT	15	22	24	15	15	13	12	21	23	21
VIRGINIA	36	56	73	111	114	187	170	382	373	321
WASHINGTON	17	29	40	86	65	108	98	180	270	226
WEST VIRGINIA	22	78	109	105	107	157	142	288	454	380
WISCONSIN	54	79	104	124	117	152	138	368	397	287
WYOMING	6	9	15	40	25	44	40	142	297	183
TOTAL	2368	3719	4679	7274	6854	9354	8597	17602	20194	16927

Table III. VOC/NOx ratio from 1900 to 1985

STATE	1900	1910	1920	1930	1940	1950	1960	1970	1980	1985
ALABAMA	4.8	3.9	3.3	5.1	4.2	2.9	1.9	1.3	1.0	1.1
ARIZONA	19.9	12.7	8.7	2.8	2.8	2.6	4.3	2.0	1.0	1.4
ARKANSAS	2.4	2.3	1.7	6.4	5.2	2.4	2.9	1.9	1.4	1.4
CALIFORNIA	12.5	7.8	6.0	1.4	1.9	2.3	3.4	2.2	1.9	2.2
COLORADO	3.5	2.3	2.2	2.7	3.1	2.7	3.9	2.1	1.1	1.2
CONNECTICUT	3.4	2.3	2.1	2.8	2.9	2.6	3.6	2.1	1.9	2.2
DELAWARE	11.2	10.4	7.7	5.7	4.4	2.5	3.8	1.4	1.0	0.9
D.C.	1.2	1.0	1.7	2.4	3.1	2.6	2.7	1.6	2.0	2.1
FLORIDA	8.0	7.1	5.0	6.9	5.5	3.8	4.4	1.9	1.6	1.7
GEORGIA	9.4	7.7	5.5	7.5	6.5	3.8	3.4	1.9	1.1	1.2
IDAHO	20.4	13.1	7.6	6.8	6.6	4.2	5.5	2.6	1.1	1.5
ILLINOIS	1.9	1.3	1.2	1.8	1.9	1.7	2.2	1.3	1.0	1.1
INDIANA	3.6	2.3	1.7	1.9	2.0	2.0	2.6	1.2	0.8	0.8
IOWA	2.0	1.5	1.6	2.7	2.7	2.2	2.6	1.6	0.9	1.1
KANSAS	2.9	1.6	2.1	1.7	2.0	1.6	2.0	1.2	0.7	0.7
KENTUCKY	6.1	4.2	3.3	3.8	3.4	2.6	3.0	1.0	0.8	1.0
LOUISIANA	11.5	9.1	4.3	1.9	1.8	1.4	1.9	0.8	0.7	0.9
MAINE	8.2	5.1	4.2	5.1	4.7	4.2	4.7	2.6	2.4	2.5
MARYLAND	5.0	3.5	3.1	2.4	2.2	2.5	3.7	1.8	1.5	1.6
MASSACHUSETTS	4.0	2.8	2.6	2.7	3.0	3.0	3.9	2.2	1.9	1.9
MICHIGAN	5.3	3.2	2.4	2.7	2.7	2.4	3.2	1.6	1.2	1.4
MINNESOTA	6.9	4.6	3.4	4.1	4.1	3.1	4.1	1.9	1.4	1.3
MISSISSIPPI	10.3	8.2	6.1	7.4	6.3	3.4	2.9	1.5	1.1	1.4
MISSOURI	5.4	3.9	3.0	3.0	3.1	2.7	3.3	1.6	1.0	1.1
MONTANA	5.6	4.2	3.2	2.9	2.9	2.4	2.7	2.1	1.1	1.0
NEBRASKA	2.4	2.0	2.1	3.0	3.1	2.6	3.3	1.7	1.0	1.0
NEVADA	17.1	7.8	6.2	1.4	2.2	2.2	2.7	2.3	1.0	1.1
NEW HAMPSHIRE	4.4	2.9	2.5	4.0	3.5	3.8	4.5	2.0	1.8	2.0
NEW JERSEY	2.5	1.7	1.5	1.3	1.4	1.9	2.7	1.9	1.7	2.0
NEW MEXICO	17.9	9.3	6.7	5.8	3.7	1.8	2.6	0.9	0.6	0.7
NEW YORK	2.0	1.5	1.5	1.9	2.1	2.2	2.8	2.1	1.9	2.2
NORTH CAROLINA	19.1	13.4	8.7	6.5	5.6	3.3	3.6	1.4	1.1	1.4
NORTH DAKOTA	4.2	2.5	2.2	3.0	3.4	2.9	3.8	1.7	0.8	0.6
OHIO	2.4	1.4	1.4	1.7	1.9	1.7	2.2	1.3	0.9	1.0
OKLAHOMA	10.5	3.9	1.9	1.8	2.8	2.1	2.1	1.3	1.0	1.3
OREGON	25.6	19.1	14.0	4.3	4.7	3.2	3.6	2.5	1.9	2.4
PENNSYLVANIA	2.5	1.4	1.2	1.6	1.9	1.8	2.2	1.5	1.0	1.3
RHODE ISLAND	1.6	1.2	1.3	1.9	2.0	2.4	3.2	2.4	2.1	2.6
SOUTH CAROLINA	17.7	12.6	8.5	7.5	5.9	3.7	4.2	1.9	1.3	1.5
SOUTH DAKOTA	4.6	3.5	3.3	3.9	3.8	3.6	4.2	2.2	1.2	1.4
TENNESSEE	11.1	8.0	6.0	5.2	4.0	2.9	3.2	1.2	0.9	1.1
TEXAS	7.2	6.1	4.3	1.4	1.4	1.2	1.7	1.0	1.0	1.0
UTAH	6.5	4.1	3.4	3.0	3.0	1.9	2.5	2.0	1.0	1.1
VERMONT	4.1	2.7	2.3	4.0	3.6	4.2	4.6	2.8	2.4	2.6
VIRGINIA	11.8	7.2	5.3	4.4	3.4	2.4	3.0	1.6	1.4	1.5
WASHINGTON	21.0	12.1	8.7	4.2	5.1	3.7	4.4	2.9	1.6	1.9
WEST VIRGINIA	5.7	1.7	1.3	1.9	1.6	1.4	1.5	0.7	0.4	0.5
WISCONSIN	5.8	3.9	3.5	3.5	3.1	2.8	3.5	1.5	1.1	1.4
WYOMING	5.7	4.5	3.2	1.4	2.8	2.2	2.9	0.8	0.4	0.6
TOTAL	5.1	3.3	2.7	2.7	2.9	2.2	2.7	1.5	1.1	1.2

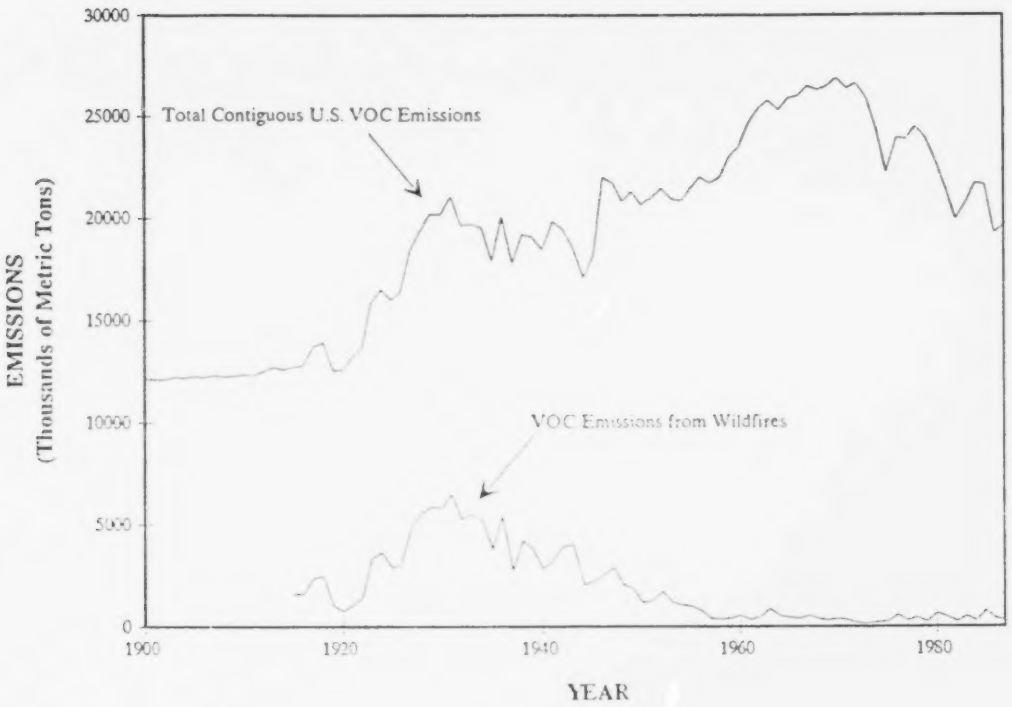

Figure 1. Total VOC emissions in the contiguous United States from 1900 to 1987.

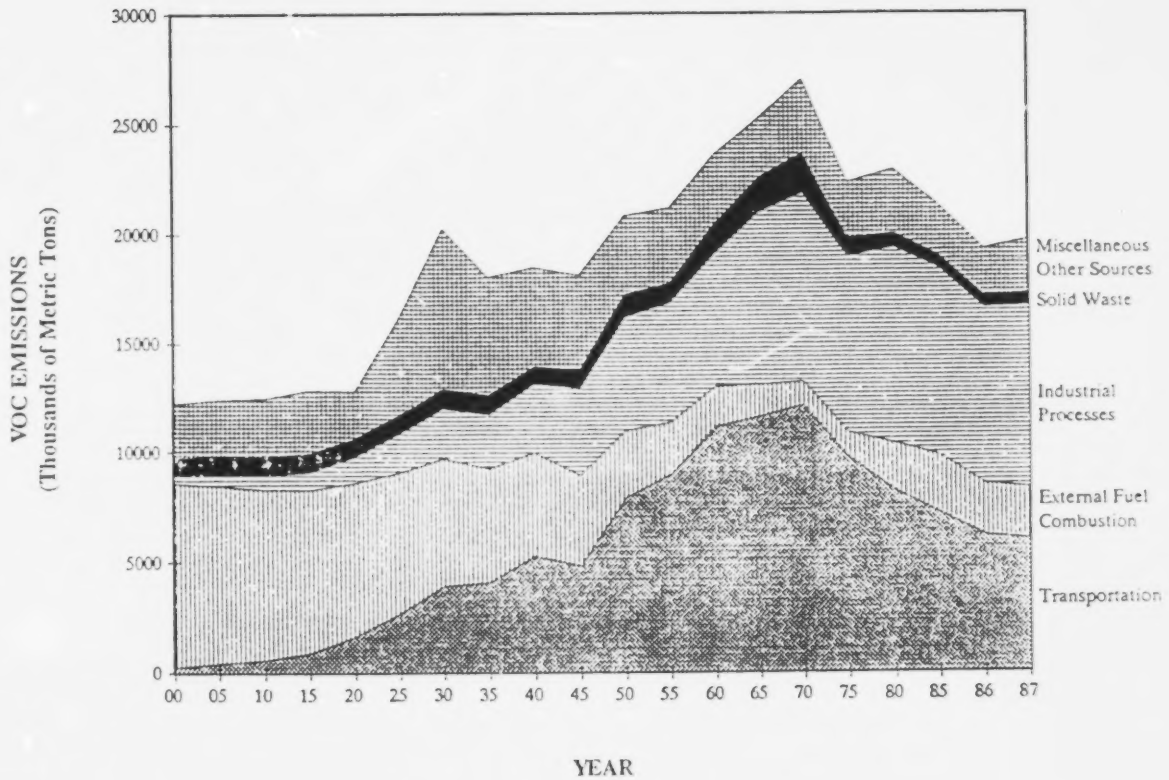

Figure 2. VOC emissions in the United States by major source category.
(Scale for 1986 and 1987 is exaggerated)

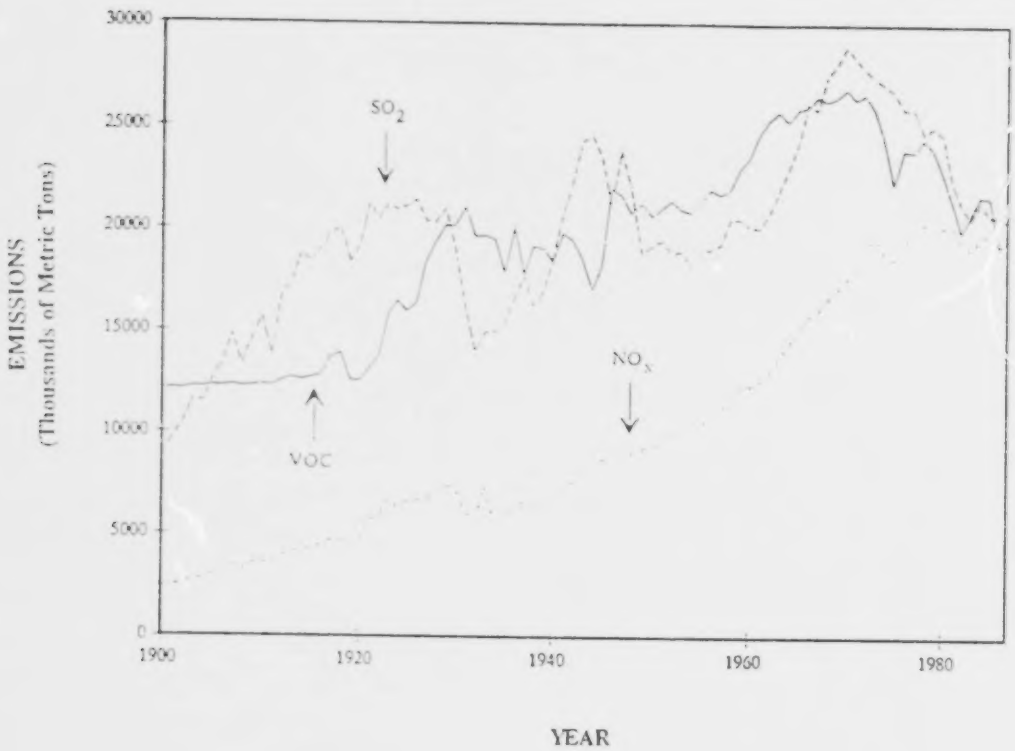

Figure 3. Comparison of total VOC, SO_2, and NO_x emissions for the contiguous United States from 1900 to 1987.

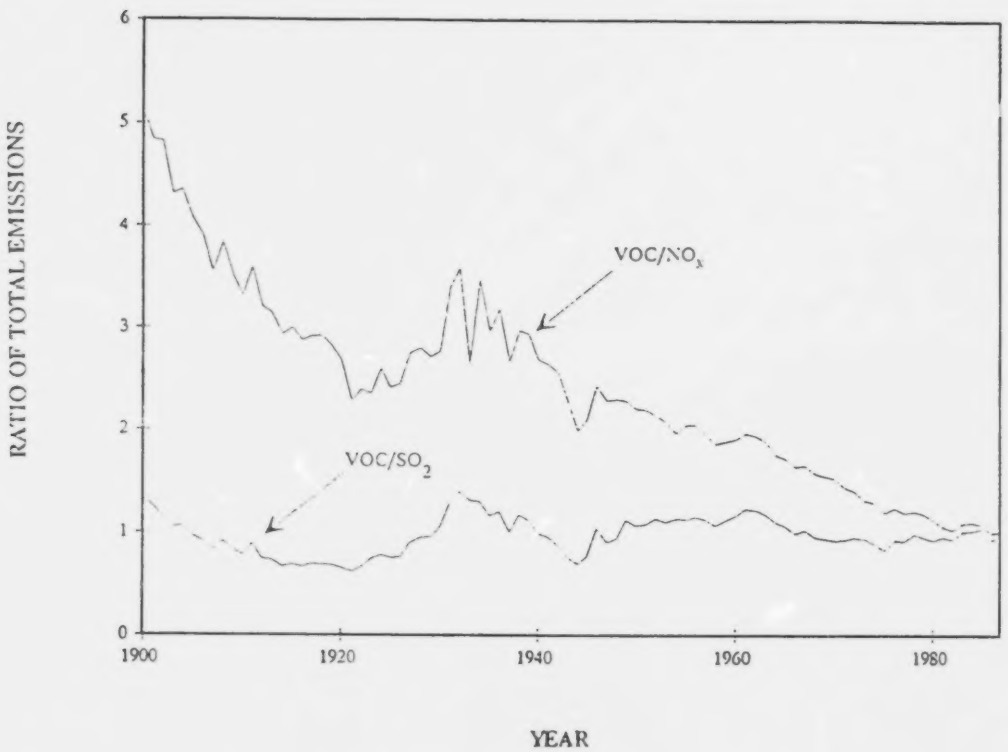

Figure 4. Ratio of VOC emissions to SO_2 and NO_x emissions from 1900 to 1987.

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